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# Plutonium Oxide Containment and the Potential for Water-Borne Transport as a Consequence of ARIES Oxide Processing Operations

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# PLUTONIUM OXIDE CONTAINMENT AND THE POTENTIAL FOR WATER-BORNE TRANSPORT AS A CONSEQUENCE OF ARIES OXIDE PROCESSING OPERATIONS

## Introduction

The question of oxide containment during processing and storage has become a primary concern when considering the continued operability of the Plutonium Facility (PF-4) at Los Alamos National Laboratory (LANL). An Evaluation of the Safety of the Situation (ESS), "*Potential for Criticality in a Glovebox Due to a Fire*" (TA55-ESS-14-002-R2, since revised to R3) first issued in May, 2014 summarizes these concerns:

"The safety issue of fire water potentially entering a glovebox is: the potential for the water to accumulate in the bottom of a glovebox and result in an inadvertent criticality due to the presence of fissionable materials in the glovebox locations and the increased reflection and moderation of neutrons from the fire water accumulation."

As a result, the existing documented safety analysis (DSA) was judged inadequate and, while it explicitly considered the potential for criticality resulting from water intrusion into gloveboxes, criticality safety evaluation documents (CSEDs) for the affected locations did not evaluate the potential for fire water intrusion into a glovebox. DOE G424.1-1B (O'Brien, 2010) states:

"For purposes of performing the Unresolved Safety Question (USQ) determination, a margin of safety is defined by the range between two conditions. The first is the most adverse condition estimated or calculated in safety analyses to occur from an operational upset or family of related upsets. The second condition is the worst-case value known to be safe."

The deficiency, by the above definition, is that otherwise well-characterized PF-4 processes have CSEDs which do not evaluate fire water intrusion, and the "worst-case value known to be safe" is therefore not known. According to the DSA, LANL's criticality safety program is:

"…implemented to prevent inadvertent nuclear criticality and to provide proper response to an inadvertent criticality and the Criticality Safety Program is a credited Safety Management Program in the technical safety requirements (TSRs). Limits and controls (engineered and administrative) are applied to fissile material operations to ensure subcritical configurations in all normal and credible abnormal conditions whenever fissile materials are present."

Lacking process-specific CSEDs which account for the intrusion of fire suppression water into PF-4 gloveboxes, LANL engineers cannot confirm the existence of subcritical configurations. Operational restrictions continue to be enforced until further information is received. For the processing of $PuO_2$ powder as described in, for example, PMT2-DOP-091 "*Sieving, Milling,* *Blending, and Sampling ARIES Oxide Powders*," considerable mitigation is obtained via engineering and administrative controls. These factors are discussed in the section of this document titled "Oxide Processing: Procedural Parameters." Routine upsets resulting in oxide spillage during processing are summarized in the section titled "Mass Loss during Processing Operations," and the quantification and handling of spilled oxide is described in "Spill Mitigation during Oxide Processing."

Finally, we believe that the information required to revise and update the existing CSEDs necessarily includes an understanding of the transport properties of Pu oxide particles in water. The ESS considers fire suppression water entering gloveboxes via four different pathways:

- Water spray from a sprinkler actuation through an air inlet filter
- Water spray through a broken or dislodged glovebox window
- Water spray through an open glove port, and
- Water spray through an open glovebox feed-through.

A number of mitigating factors for each ingress pathway are considered in the ESS. Since the ESS was issued, a number of additional controls which consider a fully-flooded glovebox environment have been codified and added as revisions to existing CSEDs in an effort to ensure that "operations involving fissionable material remain subcritical in such an upset condition" (ESS, page 7).

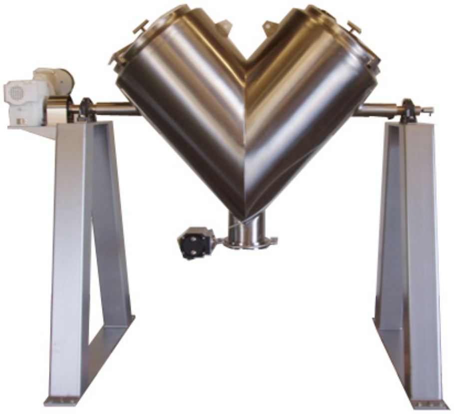
However, fire suppression water accumulation in a glovebox through a breached upper glove port, or through a broken or dislodged glovebox window, remains a concern. For example, a seismic event, by itself, could knock a glovebox off of its stand, possibly causing an unfavorable geometry from a criticality safety perspective. Not all glovebox stands in PF-4 currently meet Performance Category (PC)-2 seismic criteria, which is a known vulnerability in the DSA. If, during the same event, a glovebox window cracks, dislodges or otherwise misaligns, then a post-seismic fire could permit water ingress. Appropriate controls and bounding conditions have not yet been formulated for this particular combination of seismically-inadequate glovebox stands with CSEDs that inadequately address possible water ingress.

Part of the problem is that, in a worst-case scenario, the water so accumulated may have no way to exit the glovebox. Additionally, if the water was able to flow through a different breach, what would be the fate of the entrained radioactive solids? The behavior of ARIES $PuO_2$ powders and aggregates in water is addressed in the section below titled "Properties of ARIES $PuO_2$ Powders Relevant to Material Transport during a Catastrophic Event." In this section, settling velocities and other parameters are estimated for $PuO_2$ particulates using equations from published literature on sediment transport.

## Oxide Processing: Procedural Parameters

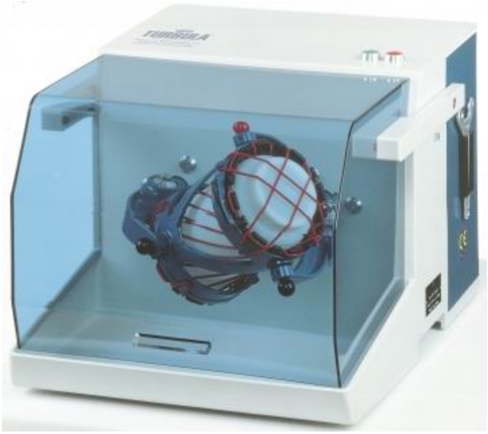
Oxide processing comprises a variety of actions taken to size-reduce, homogenize, and sort as-received $PuO_2$ aggregates in order to comply with Federally-mandated restrictions on the storage and shipment of actinide-bearing materials (e.g., DOE-STD-3013). Historically, a number of LANL programs have used a similar suite of techniques to accomplish these goals. As-received oxide is typically screened by sieving. Sieving may be accomplished by hand-shaking or tapping sieve stacks, or by placing the loaded sieve stack on an automatic shaker with a timer. For the Advanced Recovery and Integrated Extraction System (ARIES) Program, automatic sieving is performed for one hour. After sieving, the oversized material may be size-reduced in ball or rod mills. The ARIES Program uses rod milling to size reduce the oxide, with ball milling utilized for only the most refractory material. Depending on the nature of the material to be processed, and the requirements of the customer, the sieve / mill cycle may be iterated a number of times until a predetermined percentage of the material pass through a sieve having a certain mesh size. Milling duration is one hour or more.

Figure 1: Single-axis V-Blender



After the sieve / mill cycle is complete, oxide is homogenized using a small-capacity powder blender. In the past, LANL has utilized one-axis V-blenders (Figure 1) to perform homogenization. Over the last decade or so, however, the blender of choice for $PuO_2$ processing operations in PF-4 has been the 10 kg Turbula © T2F Blender (Figure 2). The Turbula T2F blender utilizes a dedicated stainless steel blending vessel, or jar, which is immobilized in an elastic mesh basket. The basket is mounted inside of an interlocked enclosure, and sits above a cast-iron tray. The largest-sized jar, which can hold up to 8.8 kg, or 2 liters, of material, is then spun on 3 axes for a predetermined amount of time. T2F-compatible blending jars are available in a variety of sizes, including 1L, 0.5L and 0.25L.

Figure 2: Turbula T2F blender used for ARIES processing.



Homogenous lots which are greater than the volume (or weight) capacity of a single, fully-charged 2L jar (e.g. 8 kg for ARIES $PuO_2$) can be created by cross-blending. In cross-blending, the entire lot is distributed between two or more jars. The contents of each jar are homogenized and then split in half. For a two-jar cross-blend, half of the contents of jar 'A' and half of the contents of jar 'B' are combined in one jar while the other jar received the other halves. Both jars are then re-homogenized. In ARIES processing, as codified in PMT2-DOP-091, cross-blending is iterated 2 to 3 times. Each homogenization takes ~30 minutes (Mason et al., 2003).

Once completely cross-blended, the material is sampled for analysis. Many programs require a simple scoop sample. ARIES requires sampling using a coring device or sample thief. For ARIES, the initial sample of 250 grams is split into smaller aliquots using a rotary riffling device such as the Fritsch Laborette 24/27. Either two 4.5 gram samples, or a 1 gram and a 2 gram sample are sent to LANL's C-AAC group or to the Savannah River Site for chemical and isotopic analyses. Particle size characterization requires a 25 gram sample, while thermal analysis requires two samples weighing between 3 and 5 grams. Bulk and tapped densities are collected on samples between 150 and 200 grams.

Figure 3: ARIES convenience can.

After sampling is complete, the fate of the fully-processed material depends on programmatic goals. Some programs merely opt for vault storage in a slip-top container which is bagged out and placed inside of a SAVY or Hagen container. By contrast, ARIES material must be packaged and sent to the Savannah River Site for further processing. The first step in packaging is to transfer the processed material to an ARIES convenience container (Figure 3) or inner can. The oxide is spooned by hand or poured into the inner can. These containers have a specified weight limit, and they are filled to meet that weight limit. Even so, they are only ~60-75% full when complete, depending on the bulk density of the granular material. When full, temporary lids are placed on the convenience cans. Each filled, lidded convenience container is placed into a purpose-built transfer container, which is sealed and sent to the Packaging area (located in a different PF-4 room) for crimping and welding.

Every lot contains more material than is required to fill the convenience containers. This fully-processed 'leftover' material is coded 'UPOPLOTxxxx-LO' ('xxxx' corresponds to the lot number) and stored until it can be incorporated into a future lot. Historically, the small amount of ARIES material that was resistant to milling and remained as particles larger than 180 micrometers (μm) in diameter was placed into a separate container, coded 'UPOPLOTxxxx-OVR,' and sent to the vault. Ultimately, the 'OVR' material was size-reduced to <180 μm by ball-milling and incorporated into future lots. Subsequently, the 'OVR' oxide was ball-milled *in situ* and incorporated into the 'LO' material without being transferred to the vault.

## Mass Loss during Processing Operations

There are numerous opportunities for $PuO_2$ powder spillage during all phases of ARIES oxide operations. The most frequent based on 3+ years of ARIES Oxide Processing operations are tabulated below (Table 1). Currently no statistics exist to quantify the frequency of oxide spillages or the weight of material lost during each spill. However, based on the data presented in Table 1, 'normal' inadvertent spillage during ARIES oxide processing will vary from less than 20 to over 160 grams, assuming 4 incoming oxide containers, 3/2 sieve/mill cycles, 2 blend cycles, 3 riffling cycles, transfer to 3 convenience cans, one instance each of riffler overflow and material impaction within the sampling thief, and no convenience can upsets.

Many vessel-to-vessel transfers are conducted with the receiving vessel inside of a clean catch pan, and the material so captured can be returned to the lot if it is free of foreign material. Material spilled onto the glovebox floor or mixed with visible foreign matter is considered contaminated and must be set aside in a designated 'sweepings' container; typically a stainless steel slip-top dressing jar. The sweepings from each lot are coded on LANMAS as 'UPOPLOTxxxx-SWP.' All sweepings (SWP) are accounted for in LANMAS. The total net weight of the oxide sweepings generated during the processing of each ARIES lot (UPOPLOT) is tabulated in Table 2. If all of the data in Table 2 is averaged, the mean mass of sweepings per ARIES oxide lot is 98.4±409.3 grams. If one exceptionally large SWP is not considered (UPOPLOT0036-SWP at 3036.8 g), the average mass in each SWP over 53 completely processed ARIES lots reduces to 42.7±39.1 g. This is well below 0.5% by weight of a typical ARIES lot.

A small amount of oxide from each lot is not recoverable via conventional means, such as sweeping. A certain fraction physically adheres to the surfaces of the processing equipment, to glovebox gloves, and to glovebox surfaces. A proportion of the finest, dust-sized fraction is entrained within the glovebox air filtration system and held up in the HEPA filters. HEPA filter changes occur every 2 to 3 years, and yield measurable quantities of mixed particulates. Unaccounted-for losses are noted and reported to SAFE-4 (LANL's MC&A organization).

Catastrophic spills during ARIES oxide processing are very rare, with only one unusual occurrence taking place during 3+ years of operation. During processing of UPOPLOT0036, the top of the mill vessel became loose during milling, and the oxide spilled out on to the roller mill and glovebox floor. This was noticed immediately by workers (though too late to stop the mill or salvage the material) and the spilled oxide was reclaimed as sweepings on LANMAS. Work was paused; the incident was reported immediately to both SAFE-4, to line supervisors, and to ARIES Program management. In this instance, processing was resumed and proceeded normally as the remaining material was sufficient to form a complete lot. A new milling vessel with a more secure lid locking mechanism was being fabricated at the time of this incident. The old vessel was decommissioned shortly thereafter, and processing has continued without incident using the new milling vessel.

**Table 1**: Actions during operations described in PMT2-DOP-091 during which spillage or mass loss of $PuO_2$ commonly occurs. Note that each lot may require several iterations of the sieve / mill sequence.

| **Action** | **Spill-likely Event** | **Mitigation** | **Estimated Mass Loss (g) per action** |
|---|---|---|---|
| Receiving Material – opening containers from DMO | $PuO_2$ powder often trapped between the outer edge of the slip top can & the lid | Perform inside of clean catch-pan & return spillage to container. | 0.5 - 2.0 |
| Sieving | Transferring $PuO_2$ to & from sieve stack: spillage and fine powder loss | Perform inside of clean catch-pan & return spillage to container. | 0.5 - 10.0 |
| Sieving | Nested sieves get stuck together & must be forced apart | Ensure sieves are clean and easily disassembled before use. Perform inside of clean catch-pan & return spillage to container. | 0.5 – 10.0 |
| Sieving | Powder hold-up on surfaces, granules trapped in mesh | Clean sieves after run. Typically this material cannot be recovered. | 0.5 – 1.0 |
| Milling | Transferring $PuO_2$ to & from milling vessel to sieves: spillage and fine powder loss | Perform inside of clean catch-pan & return spillage to container. | 0.5 - 10.0 |
| Milling | Hold-up on inner surfaces of vessel and on rods | Clean vessel after run. | 0.5 – 5.0 |
| Blending | Hold-up on inner surfaces of blend jar | Clean jar after run (most of this material cannot be reclaimed) | 0.5 – 1.0 |
| Blending | Transferring $PuO_2$ to & from blend jar: spillage and fine powder loss | Perform inside of clean catch-pan & return spillage to container. | 0.5 – 10.0 |
| Sampling | $PuO_2$ powder lodged inside of sample thief: spills outside of catch pan. | Tap thief gently inside catch pan to dislodge powder. Perform inside of clean catch-pan & return spillage to container. | 0 – 5.0 |
| Sampling | Holdup inside of riffler | Clean inside surfaces after riffling (typically this material cannot be reclaimed) | 0.5 – 2.0 |
| Sampling | Riffling proceeds too quickly; $PuO_2$ overflow during transfer from feeder to rotary unit | Monitor feed rate / control feed rate by loading incrementally. | 1.0 – 10.0 |
| Transfer to convenience can | Transferring $PuO_2$ to can from blend jar: spillage and fine powder loss | Perform inside catch pan | 1.0 – 10.0 |
| Moving full convenience can across glovebox | Can topples or is upset during move and spills oxide on the floor. | Always performed with a temporary lid on the can. Push the can gently & slowly, an inch or 2 from the bottom. (Zero occurrences to date) | 10.0 - 1000 |

**Table 2**: Net weight of sweepings (SWP) from each ARIES Lot.

| **LOT #** | net wt. $PuO_2$ in SWP (grams) | **LOT #** | net wt. $PuO_2$ in SWP (grams) | **LOT #** | net wt. $PuO_2$ in SWP (grams) |
|---|---|---|---|---|---|
| **1** | no data | **19** | 51.9 | **37** | 70.0 |
| **2** | 33.5 | **20** | 33.4 | **38M** | 20.5 |
| **3** | 28.1 | **21** | 83.8 | **39** | 38.2 |
| **4** | 4.9 | **22** | 59.5 | **40** | 54.1 |
| **5** | 30.9 | **23** | 12.8 | **41** | 27.5 |
| **6** | 7.3 | **24** | 74.9 | **42** | 19.3 |
| **7** | 55.4 | **25** | 66.3 | **43** | 25.1 |
| **8** | 13.3 | **26** | 72.1 | **44** | 47.0 |
| **9** | 32.9 | **27** | 24.6 | **45** | 30.8 |
| **10** | 21.7 | **28** | 62.7 | **46** | 28.3 |
| **11** | 40.6 | **29** | 105 | **47** | 43.5 |
| **12** | 18.9 | **30** | 25.3 | **48** | 47.3 |
| **13** | 25.9 | **31** | 51.8 | **49** | 12.7 |
| **14** | 18.4 | **32** | 46.3 | **50** | 36.7 |
| **15** | 34.6 | **33** | 14.5 | **51** | 11.3 |
| **16** | 37.8 | **34** | 229.2 | **52** | 13.5 |
| **17** | 177.5 | **35** | 20.5 | **53** | 26.8 |
| **18** | 0 | **36** | 3036.8 | **54** | 8.7 |

## Spill Mitigation during Oxide Processing

ARIES oxide processing, as formalized in PMT2-DOP-091, comprises all of the activities described above (except for HEPA filter changes), including receiving material, sieving, milling, blending, sampling, placement into convenience containers, and transfer to Packaging. The amount of material that can be sieved, milled, blended, sampled, or transferred at any one time is procedurally restricted (Table 3). Furthermore, the procedure requires that processing be performed only on oxide lots which have been specified on a formal, signed ARIES Blend Sheet. The Blend Sheet identifies the constituent oxide batches that comprise the lot, identifies their net weights, and must be reviewed and signed by MET-1 engineers, Quality personnel, SAFE-4 (Materials Control and Accountability), and NCO line managers before the material can be transferred to the Oxide Processing area. In addition, spilled material is not likely to accumulate and remain in a particular location indefinitely. Processing operators are required to clear the processing glovebox of all remnants of the previous lot before the material can be transferred in. This prevents the occurrence of cross-contamination between lots as well as overmass violations.

When not in process, the $PuO_2$ is procedurally required to be held in a closed slip-top container, with the lid circumferentially taped to the container body using designated yellow vinyl tape. All processing activities are conducted with the material in covered, lidded or sealed containers. For blending, the container is also taped shut. Thus, the opportunity for material loss is procedurally limited. Furthermore, the extent of material loss during normal operations, even during a worst-case event, is limited by engineered restrictions (Table 3) such as container capacity, fully-charged container weight, interlocks, and the use of catch pans.

In light of the new criticality safety measures implemented in the wake of the June 2013 pause, oxide processing activities were required to cease. There still remain questions as to the potential risks of oxide processing operations vis-à-vis nuclear criticality, particularly in the case of catastrophic glovebox flooding. However, a number of bounding conditions that enhance the safety of oxide processing operations are already in place and should be credited. Furthermore, the amount of free, unattended, non-containerized $PuO_2$ powder in a glovebox at any given time is in the 10s to 100s of grams, and is well-known and well-characterized.

We can propose a number of new procedural restrictions on oxide processing capabilities that can be included in a revised CSED. With these added measures, oxide processing may proceed in a manner that meets or exceeds the new standards for safe operations in PF-4:

- Reduce the lot sizes required by the IDC.
- Reduce criticality limits (prior to the 2013 pause in operations, up to 20 kg Pu in oxide / 10 kg per container was permitted per NCS-CSED-11-176).
- Place oxide in Hagan or SAVY containers when not in process (sieving / milling / blending / sampling / riffling).
- Use smaller, lower volume blending jars, reduce amount of material to be blended at any one time.
- Reduce the mass of oxide to be sieved and milled at any one time.
- Credit $^{240}Pu$ content to reduce constraints on fissile mass.
- Add PC2-compliant seismic stabilizers to the glovebox stand.

If some, or all, of the additional restrictions and credits are imposed upon Oxide Processing operations in PF-4 gloveboxes, some question may still remain as to the criticality-relevant behavior of $PuO_2$ particulates in a flooded glovebox scenario. Though it is unlikely, the possibility for a glovebox flooding event must be addressed in the process-specific CSEDs. Without an understanding of the form and fate of $PuO_2$ powder in a flooded glovebox, we cannot account for the intrusion of fire suppression water into PF-4 gloveboxes, and LANL engineers cannot confirm the existence of subcritical configurations. Operational restrictions shall continue to be enforced until further information is received. The objective of the next section of this document is to provide a technical and numerical basis for the understanding of $PuO_2$ powder behavior in a flooded glovebox.

**Table 3**: Current (as of Nov. 2014), non-ESS procedural and engineered restrictions relevant to spill size reduction during oxide processing operations.

| ACTION | Procedural Restriction in PMT2-DOP-091 | Engineered Restriction |
|---|---|---|
| Material present in FMO | "Ensure that no process material (-LO, -OVR, -SWP) from the previous lot remains in the processing glovebox…" (p. 19)<br>"When not being used in a process (sieving, milling, blending, sampling, transferring from one container to another), all containers, cans, vessels, jars, etc. containing SNM shall be lidded or covered. The lid shall then be taped circumferentially to the container body using yellow vinyl tape." (new text to be added during final revision) | none |
| Pouring from one container into another | none | Slip top capacity is ≤4.5 kg for ARIES oxide<br>Pouring done with secondary confinement (receiving can inside pan) |
| Sieving | Sieving restricted to <2500 grams of $PuO_2$ per run (p. 21).<br>Sieve stacks are required to be lidded during operation | Sieve shaker platform has a ~0.25" lip that secures sieves from bottom<br>Sieve stack attached to shaker platform w/a restraining strap and lid. |
| Milling | Milling batches restricted to <4 kg of $PuO_2$ (rod mill) or <1 kg of $PuO_2$ (ball mill) (p. 23)<br>Procedure permits tightening of lid seal with a wrench | On/off switch external to glovebox permits mill to be turned off in event of leakage, slippage, etc.<br>Lid closure design does not permit the lid to fall off even if it becomes loose during operation. |
| Blending | Blending restricted to 75% volume capacity (for a 2L jar: ~7.5 kg @ 5.0 g/mL bulk density; 8.25 kg @ 5.5 g/mL bulk density)<br>Jar lid closure required to be taped shut before blending. | 1L, 0.5L, 0.25L jars are available for use, pending criticality evaluation.<br>Blending takes place within an interlocked enclosure<br>Unit permanently mounted on a cast iron base with a ~1" lip that catches falling particles. |
| Sampling | Procedure allows for dividing 250-gram samples into smaller samples. | Sample pouring, decanting, etc. performed inside a large catch pan. |
| Transfer to Convenience Container | Convenience container maximum capacity is 4800 grams; ~60-75% full | Sample pouring, decanting, etc. performed inside a large catch pan. |
| Manipulation of Convenience Container | Slip-top lid placed over Convenience container prior to movement and placement in transfer container<br>Transfer container sealed shut with thumbscrews before it is moved to another area | Transfer container has O-ring seals |

## Properties of ARIES $PuO_2$ Powders Relevant to Material Transport during a Catastrophic Event

Plutonium oxide, $PuO_2$, is highly insoluble in water (Machuron-Mandard and Madic, 1994). Therefore, our main concern is the physical behavior of $PuO_2$ particles in water. The most thoroughly studied aspect of the transport behavior of $PuO_2$ powders or particles concerns the suspension and resuspension of airborne respirable fines (Mewhinney et al., 1987; Kogan and Schumacher, 1993; Marlow and Cheng, 2003; Cheng et al., 2004; Nakae et al., 2005). The end user for ARIES oxide, MOX Services, has requested that LANL "…attempt to make the proportion of sub-5-µm particles as low as practical" for oxide shipped to Savannah River Site. Eventually, MOX Services requested that the volume of sub-5-µm fines be limited to less than 30%. However, this specification was made primarily to prevent clogging of their highly automated dissolution system with fines.

Very little work has been done on the suspension and transport of $PuO_2$ fines in water. A descriptive study by Panesko (1972) indicates that the interaction of loose $PuO_2$ having a bulk density of 1.9 g/cc with fire suppression water spray is dominated by the formation of a saturated surface layer on top of otherwise dry oxide. The situation changes when water droplets or a discrete stream of water contacts the oxide powder. In these situations, the particulates spatter and spread out on the underlying surface. Continued flooding causes the formation of a "protective" layer of water on top of the oxide which inhibits further agitation, depending on the force of the impinging water stream. Fines of unspecified size were observed to stay in suspension above the coarser particulates for over 24 hours. Unfortunately, Panesko's report includes nothing in the way of quantitative measurements of the time required for $PuO_2$ particles of a measured size range to settle out of a static water column.

An exhaustive report by Serne, et al. (1996) addresses possible scenarios related to potential criticality in Hanford waste tanks. The main thrust of this work assesses the likelihood of plutonium-rich solids segregating from potentially neutron-absorbing non-SNM sludge *in situ*, such that an unfavorable geometry obtains. Serne and colleagues performed quantitative fluid dynamic calculations using both one- and three-dimensional conceptual models and codes assuming that that both fissile and neutron absorbers agglomerated only with themselves to form particles whose size distributions corresponded to those measured by light-scattering and visual-settling techniques. Serne, et al. (1996) concluded that the level of segregation of fissile materials from neutron absorbers was about a factor of 2.5, while the concentration necessary to reach criticality was estimated to be about 20, based on the highest measured concentration of plutonium in tank sludge. Worst case settling calculations leading to separation factors >20 required that: 1) the plutonium constituted the finest grained particles at the highest concentration for any tank at the Hanford Site; 2) that the neutron absorbers were the larger particles in the sludge; and 3) that no interparticle agglomeration occurred. In the extreme case, all neutron absorbers settled first and the plutonium slowly settled on top to form a thin, fissile-rich layer that exceeded the 2.6 g/L safe plutonium concentration for infinite geometry. Even in this extreme case, the fissile layer had to adopt a uniform pancake shape, far below the 240 g/ft$^2$ necessary to go critical. Serne, et al. (1996) considered a wide range of possibilities whereby separation factors could be so enhanced, including tank-to-tank transfers, cascading, salt-well pumping, air-lift circulation, in-tank mixing, and jet-sluicing. They concluded that such operations would not produce fluid dynamic conditions which would lead to such large separation factors.

## ARIES Oxide Production

The Advanced Recovery and Integrated Extraction System (ARIES) produces high-purity $PuO_2$ by oxidizing Pu metal from decommissioned weapons in a mechanized Direct Metal Oxidation (DMO) Furnace (Scogin, 2001; Kolman, 2002; Zygmunt, 2004). The 1st ARIES Demonstration was performed during the late 1990s using a furnace, DMO-1, adapted from the HYDOX furnace (Colmenares et al., 1996)  which oxidized metal in a rotating, perforated basket within a controlled, $O_2$-rich atmosphere at ~600°C. Oxide spalls off reacting metal surfaces, falls through the perforations and into a fritted quartz tube which served multiple functions: gas inlet, oxide reservoir, and calcination furnace. However, only a few of the oxide batches were calcined to ~600°C. The same setup was used for the 2nd ARIES Demonstration during the early 2000s.

All ARIES oxide production (UPOPLOT), and all ARIES testing and development from 2009 to the present time has taken place in the DMO-2 Furnace, an improved version of the DMO-1 furnace (Kolman, 2002; Zygmunt, 2004). The oxidation sector of DMO-2 retains the same basic features employed in DMO-1. Oxidation is maintained between 475 and 575°C in an optimized helium – oxygen mixture (75% $O_2$ / 25% He) which flows into the reaction zone at 2.0 L/ min. The total amount of $O_2$ available for Pu oxidation exceeds that needed to produce stoichiometric $PuO_2$. The DMO-2 furnace design utilizes a screw calciner, unlike DMO-1. Oxide chips and powder from the basket are collected and calcined for 130 to 135 minutes at 950-1040°C in the $O_2$-He atmosphere.

Compared to the oxalate-precipitated $PuO_2$ produced by MR&R and MOX Pu Polishing, ARIES oxides are characterized by their high bulk and tapped densities, low surface area, very low moisture content, and their distinctive tri-modal particle size distributions (Wayne 2009, 2014). Scanning electron microscopy indicates that ARIES $PuO_2$ particles possess relatively simple surface features, with irregular to prismatic shapes (Figure 4). Taken together, the high bulk and tapped densities, low surface area and physical appearance of ARIES oxides indicates that they are appropriately modeled as impermeable, non-porous particles.

## $PuO_2$ Particles in Water: Sink or Swim?

Stokes' Law can be used to describe the terminal or settling velocity of impermeable spherical solid particles immersed in a liquid medium by setting the combined forces of drag and buoyancy ($F_b$) equal to gravitational force ($F_g$) and solving for velocity ($V_s$, in m/s), where R is the particle radius (m), $g$ is gravitational acceleration in m/$s^2$, $\rho_p$ is the particle's solid density (kg/$m^3$), $\rho_f$ is the mass density of the fluid (kg/$m^3$), and $\mu$ is the dynamic viscosity of the fluid (kg /m*s).

$$v_s = \frac{2}{9}\frac{(\rho_p - \rho_f)}{\mu} g\, R^2$$

The settling velocity is strongly dependent on particle size and density. Viscosity, in the denominator, varies as a function of temperature. Stokes' Law applies only to homogeneous, smooth, non-interacting spherical particles in a homogeneous fluid. Stokes' Law also applies only to particles having a low Reynolds number (Re):

$$\mathrm{Re} = \frac{\text{inertial forces}}{\text{viscous forces}} = \frac{\rho \mathbf{v} L}{\mu} = \frac{\mathbf{v} L}{\nu}$$

...where $\rho$ is the mass density of the fluid; **v** is the mean velocity of the object relative to the fluid; L is a characteristic length, $\nu$ is the kinematic viscosity of the fluid ($m^2$/s) and $\mu$ is the dynamic viscosity. Stokes' Law applies under laminar flow conditions where viscous forces are dominant, and are characterized by smooth, constant fluid motion. For quartz particles in water at 20°C, Stokes' Law begins to break down for grains having diameters ≥0.1 mm (Ferguson and Church, 2004) as the turbulent drag caused by the wake behind larger grains becomes significant.

The determination of the settling velocity of suspended particles is also a fundamental variable in the geosciences; specifically in the field of physical sedimentology. Sedimentologists have developed a number of highly sophisticated models to quantify settling velocities based on experimental observations of particles having various diameters, densities, and shapes in a variety of environmental conditions. The model to determine non-Stokes settling velocities proposed by Ferguson and Church (2004) is computationally simple and corresponds extremely well to the experimental data:

$$w = \frac{RgD^2}{\sqrt{C_1 \nu + (0.75 C_2 RgD^3)}}$$

...where $R$ is the submerged specific gravity of the particle (defined as $[\rho_p - \rho_f] / \rho_f$), $g$ is the standard acceleration due to gravity (=9.80665 m/$s^2$), $D$ is particle diameter, and $C_1$ and $C_2$ are coefficients which account for particle non-sphericity and drag coefficient. For spherical particles, $C_1 = 18$, and $C_2 = 0.4$; for highly irregular particles, the values become 24 and 1.2, respectively. For particles of nominal diameter, as in the case of the model results presented below, $C_1 = 20$, and $C_2 = 1.0$.

Figure 4: SEM backscatter image of typical ARIES oxide.

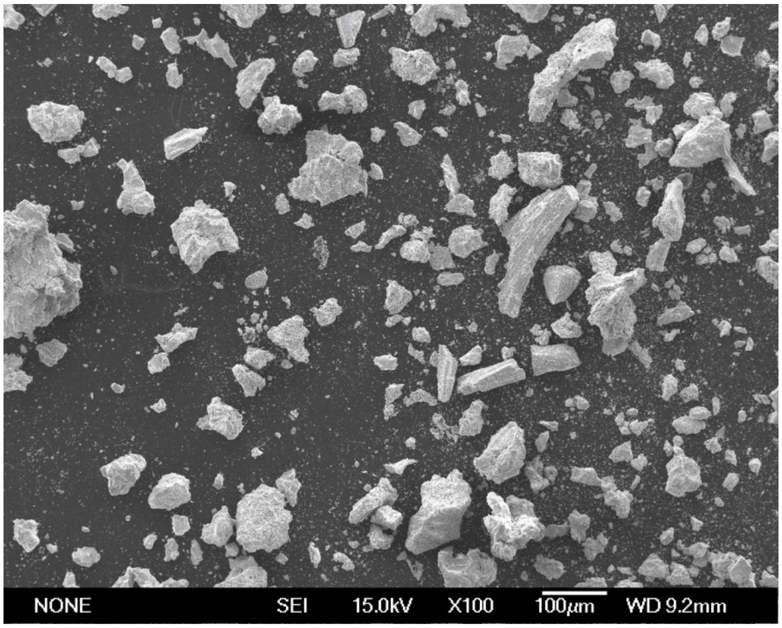


Figure 5 depicts the settling velocities (in m/s) for different-sized particles of $PuO_2$ (density = 11.5 g/cc) and quartz (density = 2.65 g/cc) using both Stokes' Law (solid line) and the formula from Ferguson and Church (2004), assuming water temperature =298K and a kinematic viscosity of $9.025 \times 10^{-7}$ $m^2/s$. Figure 5 indicates that Stokes' Law breaks down for $PuO_2$ particles of about 50 µm in diameter; with significant deviations occurring above ~100 µm. Stokes' Law is the *less* conservative case, as it does not take non-sphericity and drag coefficient into consideration. For a water-filled glovebox, the "worst case scenario" would be one in which all $PuO_2$ particles are suspended uniformly for an indefinite time period. However, the maximum residence time for $PuO_2$ particles of a given diameter in suspension can be calculated given the depth of the water in the glovebox divided by the settling velocity.

Figure 6 is a plot of the time (in hours) required for $PuO_2$ particles to settle out of a static column of water from heights of 1.0, 0.5 and 0.1 meters. According to Stokes' Law, a $PuO_2$ particle 1 µm in diameter would require approximately 49 hours to settle out of a 1 m column of water, half as long to travel 0.5 m, and 10% as long to travel 0.1 m. The equation of Ferguson and Church (2004) gives a near-identical result at this diameter. A $PuO_2$ particle 2.5 µm in diameter would require less than 8 hours settle out in a 1 meter-deep water column. Increasing the diameter to 10 µm, decreases the settling time to about 30 minutes and a particle 50 µm in diameter would settle out of a 1 m column in about 1.4 minutes using the more conservative approach devised by Ferguson and Church (2004).

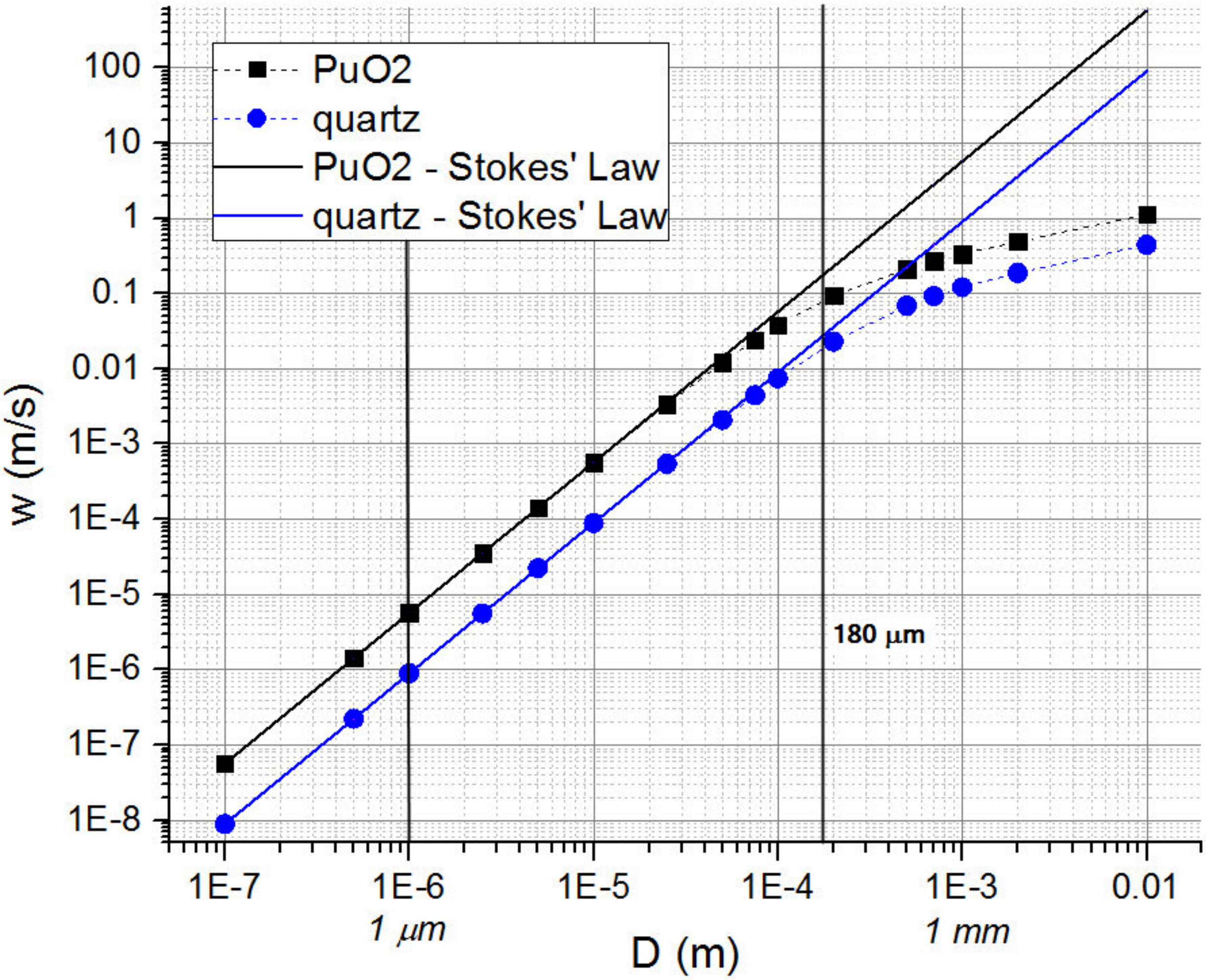
Three additional factors may complicate the scenario depicted in Figure 6. Firstly, very tiny particles (< 1.0 µm) may remain in suspension longer than predicted by gravitational settling formulas due to Brownian motion. The radius of a sphere too large to be effected by Brownian forces can be predicted by the dimensionless Péclet (Pe) number (Jonasz, 2006):

$$Pe = v_S\, a / D_{Br}$$

…where $v_S$ [m $s^{-1}$] is the terminal (Stokes) settling velocity of a particle with half-size $a$ [m], and $D_{Br}$ is the diffusion coefficient. Settling velocity ($v_S$) of a solid spherical particle in a static fluid is expressed by:

$$v_S = [g / (18\, \eta_f)]\, D^2\, (\rho - \rho_f)$$

**Figure 5: Plot of calculated settling velocities vs. particle diameter for $PuO_2$ (black) and quartz (blue) using Stokes' Law (solid line) and the formula given by Ferguson and Church (2004). T=298K, kinematic viscosity of water = 9.025x$10^{-7}$ $m^2$/s.**



…where $g$ [m $s^{-2}$] is the acceleration due to gravity, $\eta_f$ is the dynamic viscosity [kg $m^{-1}$ $s^{-1}$] of the fluid medium, $\rho$ [kg $m^{-3}$] is the density of the particle, and $\rho_f$ is the density of the fluid. The Péclet number (Pe) for a spherical particle equals:

$$Pe = 4\pi\, \Delta\rho\, g\, a^4 / (\, 3\, k_B T\,)$$

…where $\Delta\rho$ is the differential density of the particle ($\rho$ - $\rho_f$), $a$ [m] is the particle radius, $k_B$ [J K$^{-1}$] is the Boltzmann constant, and $T$ [K] is the absolute temperature of the dispersion.

The Péclet number, *Pe*, is analogous to the ratio of energy expended by the gravitational settling of a particle along a distance equal to its radius, $a$, to the thermal energy, $E_{therm} = k_B T$. If $Pe >> 1$, the effect of the Brownian motion on the particle is negligible. It follows that the critical value of the sphere of radius, $a$, that yields $Pe = 1$, is:

$$a = [3\ k_B T\ /\ (\ 4\pi\ \Delta\rho\ g\ )]^{1/4}$$

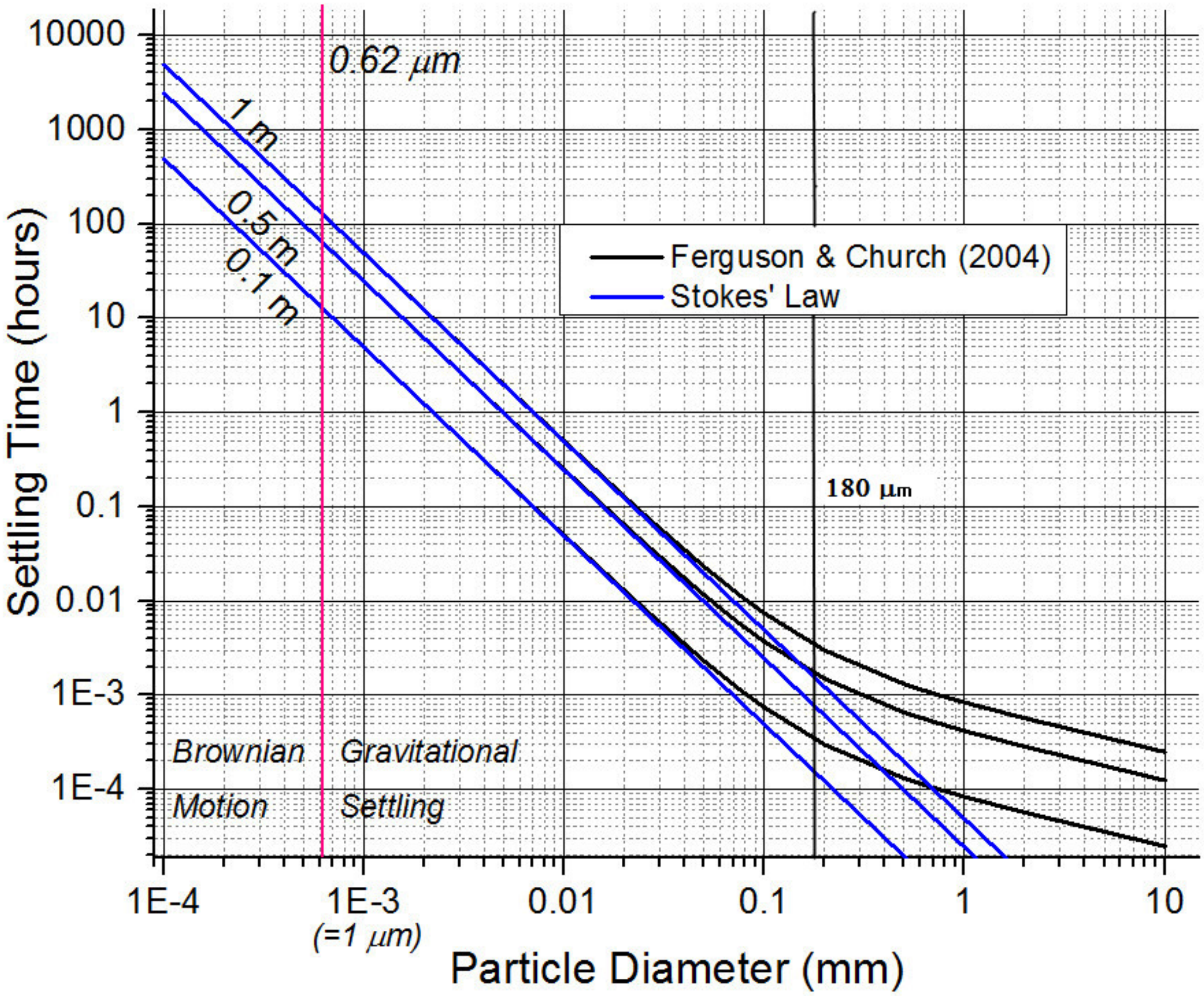
For a single polystyrene sphere ($\Delta\rho$ ~ 50 kg m$^3$) in water at 23°C, the critical radius, $a$, is 1.2 µm. For a $PuO_2$ particle ($\Delta\rho$ ~10500 kg m$^3$), in water at 25°C, the critical radius, $a$, is approximately 0.31 µm. Thus, gravitational settling will dominate the behavior of impermeable, non-porous $PuO_2$ particles having radii larger than about 0.31 µm, or diameters greater than 0.62 µm (Figure 6).

Second, at high particle concentrations, particle settling may be delayed as both hydrodynamic and particle-to-particle interactions increase in importance. This phenomenon is known as "hindered settling." The relative particle concentration ($c\ /c_{max}$) at which hindered settling becomes significant is dependent on the particles' Reynolds number (Re) and on the volume fraction occupied by the particles ($\Theta$). Using the calculation tool written by Rouweler (2013), the effect of hindered settling for spheres having concentrations of $\Theta$ = 0.1 and 0.5 reduce the free-fall gravitational settling velocity of spherical $PuO_2$ particles to approximately 60% and 40%, of the free-fall settling velocities predicted in Figure 5. A particle volume fraction ($\Theta$) of 0.75 reduces particle settling to almost zero. However, at these very high volume fractions, the particles may already be considered to be settled out of solution. We consider hindered settling will probably not be very significant at the water-to-solids ratio that would obtain in a flooded PF-4 glovebox (2.438 m l. x 1.524 m w. x 1.1176 m d.), even at incredible loadings >20 kg.

The third factor to consider is particle flocculation. It is also possible, that the observed settling velocities for $PuO_2$ particulates are biased by agglomerations formed via flocculation. Desroches, et al. [2005] have shown that such agglomerates can even form during particle size analysis, introducing significant bias to PSD measurements. In a glovebox flooding scenario, particulate agglomerates may either sink *more* rapidly, due to their increased mass and permeability, or *less* rapidly due to rafting on the surface if the particles are hydrophobic.

The tendency for particle surfaces to resist wetting, agglomerate and raft is a function of the particle's isoelectric point (IEP). The term "isoelectric point," also known as the zero point of charge (ZPC), represents the pH at which an immersed solid surface has a zero net charge. The IEP of $PuO_2$ occurs at approximately pH=9 (Kosmulski, 1997), thus $PuO_2$ particles are hydrophilic and tend not to flocculate in near-neutral aqueous media.

**Figure 6: Settling time (hours) versus particle diameter (μm) for $PuO_2$ in a static water column of 1.0, 0.5 and 0.1 meters in height. Both Stokes' Law and the settling model of Ferguson and Church (2004) are shown. The red vertical line denotes the diameter of a PuO2 particle corresponding to Pe = 1.**



Finally, settling rates may also change in response to the size and shape of the water column, the presence of a turbulent flow, and the proximity of the falling particle to the container wall. Further information on these special cases will be developed as needed.

## Particle Size Distribution of ARIES $PuO_2$

Particle size distribution analyses have been a required component of analytical data suites for ARIES and other PF-4 programs for many years. Particle size distribution data from high-purity (>85 wt. % Pu) ARIES $PuO_2$ powders and aggregates are summarized in Table 4. Note that, outside of ARIES, the physical properties, including specific surface area, bulk and tapped densities and particle size distribution, of high-purity $PuO_2$ powders originating via different processes may vary significantly (e.g., Narlesky et al., 2012).

In general, oxides created by metal burning at high temperatures results in a coarse, granular aggregate (Wayne 2014) largely comprised of particles between 180 and 1000 μm in diameter. Fines from these powders have trimodal differential particle size distribution (Wayne, 2014), as do the fully-processed ARIES oxides.

ARIES oxide (Figure 7) is generated directly from the metal in a furnace at 475 – 575°C in an oxygen-rich atmosphere (Wayne, 2014). This oxide was then further calcined in the same oxygen-rich environment at ~1000°C. $PuO_2$ emerging from the DMO furnace, or the muffle furnace, typically has a very wide particle size distribution. Wayne (2014) used calibrated sieves to characterize the particle size distribution of raw, unprocessed oxide emerging from both the DMO furnace and from a muffle furnace. During the early phases of ARIES DMO testing and demonstration, the <180 μm fraction of the raw, unprocessed oxide typically constituted <30% by weight of the entire batch, necessitating several sieve / mill iterations during processing. In August 2011, a mechanical malfunction caused DMO basket rotation to slow or cease entirely. This off-normal condition produced oxide having up to 70 weight % <180 μm fines and approximately 10 weight % (or less) coarse material >800 μm in diameter. As soon as normal DMO furnace operations were restored in September 2011, the sieve PSD of the unprocessed ARIES oxide gradually returned to its former pattern. Oxides produced by static oxidation in a muffle furnace in ambient air contained between 35.7 and 73.4 percent <180 μm fines, by weight, with little or no content above 800 μm. Milling and sieving tests demonstrate that ~20% of the >1000 μm material is eliminated after one minute of milling, with concomitant increases at smaller-diameter populations. The >1000 μm population is eliminated after 5 minutes, and after 15 minutes of milling, nearly 65% of the material passes through the 180 μm sieve. After 30 minutes, over 95% of the material passes through the 180 μm sieve.

**Figure 7: As-received oxide from the ARIES DMO Furnace (left) and processed ARIES powder after milling, sieving and blending (right).**

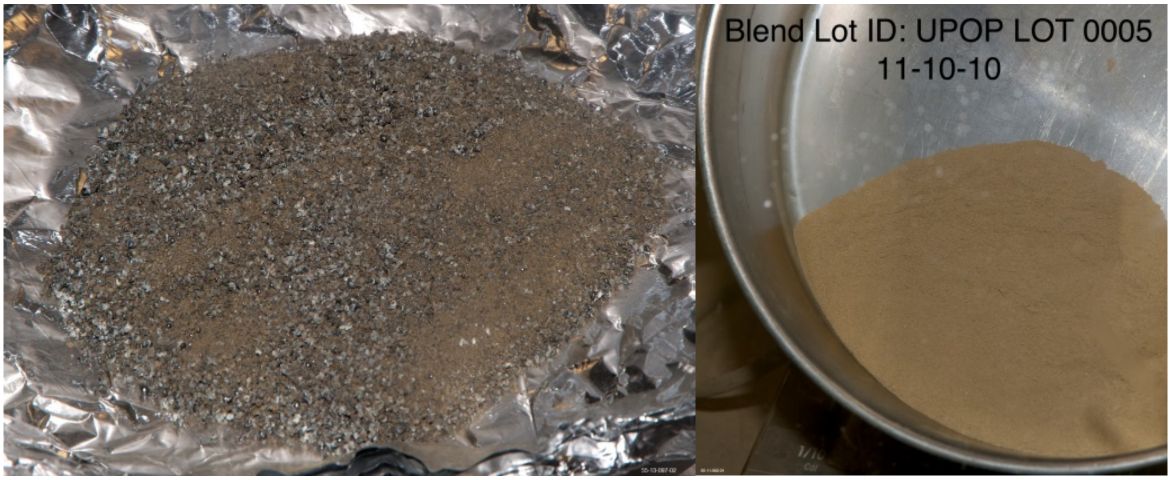
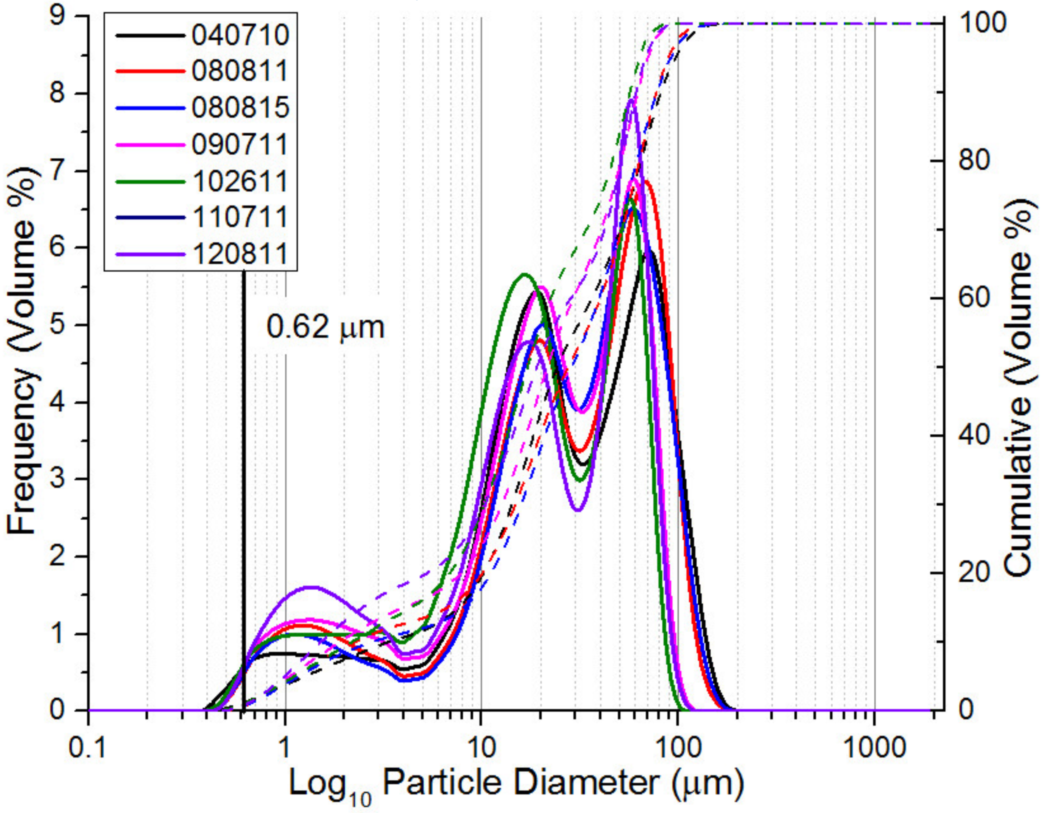


**Table 4:** Summary of particle size distribution data for high-purity ARIES $PuO_2$ powders and aggregates.

| Oxide Type | Smallest Particles Detected (μm) | Volume % Particles <1 μm | Volume % Particles <5 μm | Volume % Particles <10 μm | Number of modes | Mean PS in μm (over each mode) | Median PS in μm (over each mode) |
|---|---|---|---|---|---|---|---|
| ARIES <180 μm fraction (not processed) | 0.48±0.04 | 4.7±1.1 | 15.2±2.9 | 23.8±4.5 | 3 | 1.54 / 16.1 / 60.1 | 1.32 / 15.4 / 57.3 |
| ARIES MFTP <180 μm fraction (not processed) | 0.49±0.03 | 3.8±0.9 | 17.0±3.7 | 26.7±4.7 | 3 | 1.72 / 15.2 / 56.1 | 1.55 / 14.6 / 54.2 |
| ARIES (processed) | 0.50±0.05 | 4.7±2.3 | 21.4±4.4 | 34.0±4.4 | 3 | 1.74 / 14.3 / 52.2 | 1.56 / 13.4 / 50.5 |

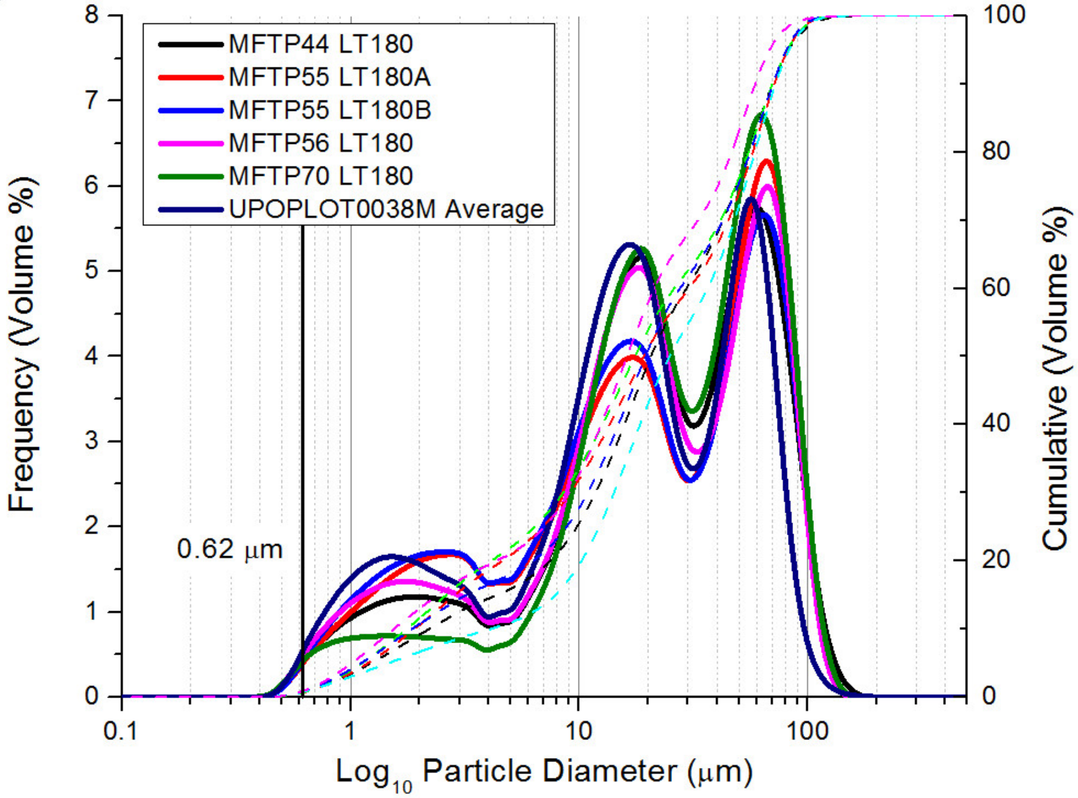
*Note: All samples ultrasonically agitated for 10-15 seconds.*

Figure 8: PSDs of <180 μm fraction from unprocessed ARIES $PuO_2$



Laser diffraction particle size distribution analyses (Figures 8-10) were performed only on oxide material that passed through a 180 μm sieve. Thus, the data for unprocessed ARIES oxide and unprocessed MFTP oxide constitutes the PSD of ~25% and ~75% by weight, respectively, of the total unprocessed mass; the remainder being >180 μm in diameter. For processed (sieved, milled, and blended) ARIES oxide, 100% of the material is <180 μm in diameter.

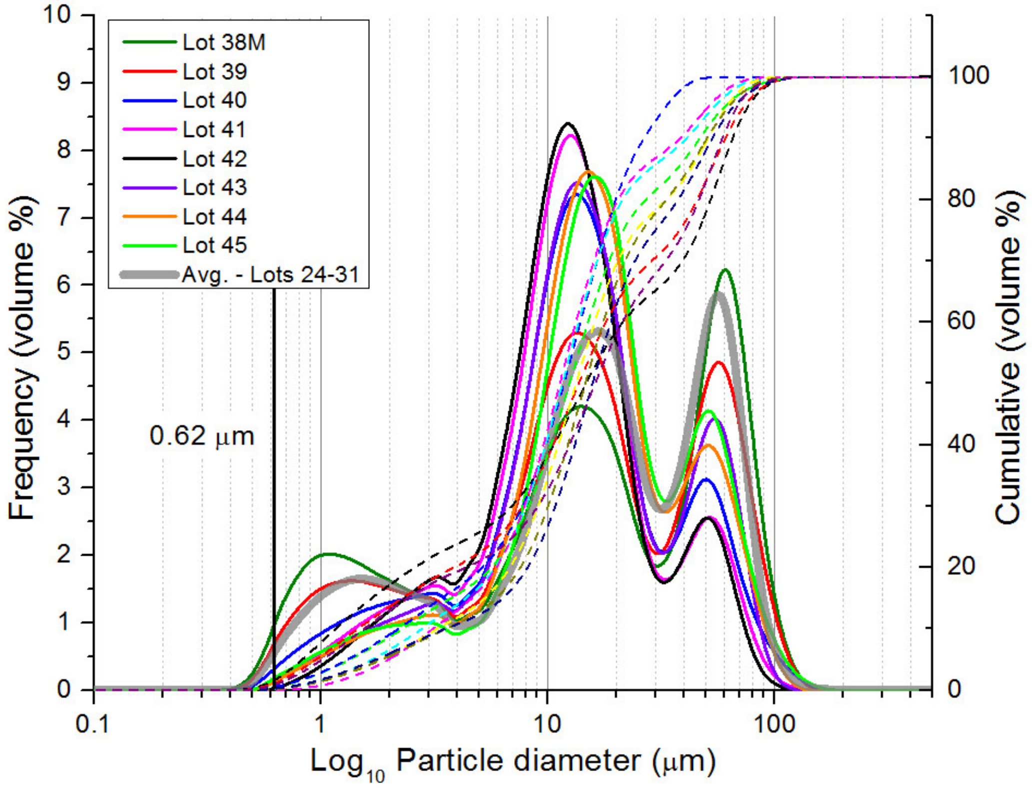
Figure 9: PSDs of <180 μm fraction from unprocessed MFTP $PuO_2$



Particle size distribution measured using laser diffraction is represented as volume distributions across a series of particle size bins; similar to--but much smaller than--one would use in a sieve analysis. In other words, the plots show the contribution of each particle size bin to the overall volume of the sample. It is important to remember that a number of tiny particles will occupy a much smaller volume than the same number of larger particles. For ARIES $PuO_2$ powders of all types (Figures 8-10), 75% to 90% of the total particle volume comes from particles between 5 and 100 μm in diameter. For the PSD plots, it is also important to distinguish the differential frequency distribution of the particles in each particle size bin (solid lines, left-hand Y-axis) from the cumulative percentage of the particle sizes over the whole sample (dashed lines, right-hand Y-axis). Both parameters are stated in terms of volume percent (volume %). The differential frequency distribution (left-hand y-axis) corresponds to a probability density function and refers to the fraction of particles (as volume %) represented within a given particle size bin. The cumulative distribution (right-hand y-axis) simply adds the population in each particle size bin of the differential frequency, and refers to the fraction (as volume %) of undersize particles that occupy a single particle size bin over the entire sample.

Table 4 also shows the range of values obtained for the smallest diameter particle in each sample. The cumulative volume percent of fine particles in the size fractions below 10 μm, 5μm, and 1 μm varies slightly for the different classes of ARIES oxides.

Figure 10: PSDs of processed ARIES UPOPLOT $PuO_2$ (all <180 μm)



For $PuO_2$ particles in standing water at 20°C, the only variable required to solve either Stokes' Law or the settling velocity equation given by Ferguson and Church (2004) is particle diameter (or radius). Based on the Péclet radius, *Pe*, calculated for a solid, impermeable $PuO_2$ particle, we predict that the gravity settling of particles ≤0.62 μm in diameter will be delayed by the effects of Brownian motion. The PSD data in Table 4 and Figures 8-10 indicate that the quantities of particles in the size range near the *Pe* for $PuO_2$ (0.31 μm or 0.62 μm in diameter) are between 1 and 2% by volume for ARIES oxides. In other words, 1% to 2% of a given volume of ARIES oxide powder will consist of particles having radii below the critical length at which Brownian motion dominates over gravitational settling. The behavior of the remainder of the $PuO_2$ in a glovebox flooding scenario will be dominated by gravitational settling, as shown in Figures 5 and 6. Table 5 summarizes the results of the settling calculations shown graphically in Figure 6. The majority of the oxide will settle out on to the glovebox floor in less than a minute to several hours, with the fines ≤5 μm requiring at least 2 hours to fall 1 meter in a static water column. Fines between 0.62 and 1 μm in diameter will require ~48 to 100 hours to settle out of a 1 meter water column.

**Figure 11: PSDs for $PuO_2$ generated by the passive oxidation of Pu metal in glovebox air at ambient conditions.**

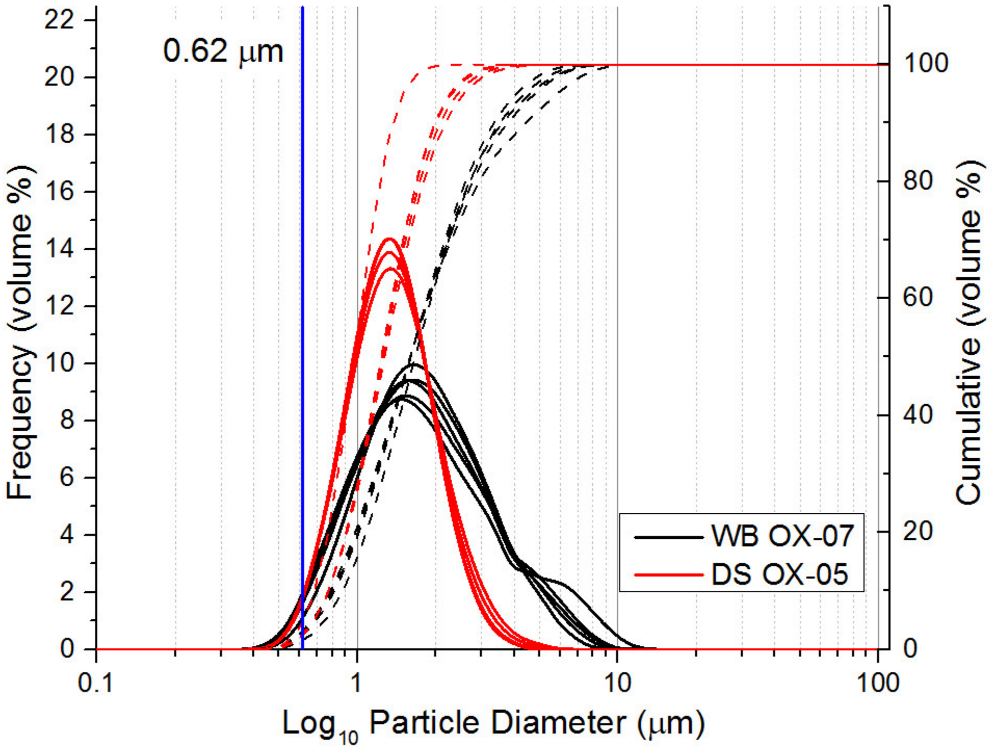


**Table 5: Results of particle settling models for $PuO_2$ particles in 1.0, 0.5, and 0.1 m of standing water.**

| Particle diameter (mm) | Pe | Ferguson & Church (2004) | | | Stokes' Law | | |
|---|---|---|---|---|---|---|---|
| | | *Time (h) for 1 m* | *Time (h) for 0.5 m* | *Time (h) for 0.1 m* | *Time (h) for 1 m* | *Time (h) for 0.5 m* | *Time (h) for 0.1 m* |
| 0.0001* | 7.18E-04 | 4869 | 2435 | 487 | 4869 | 2435 | 487 |
| 0.0005* | 0.449 | 195 | 97.4 | 19.5 | 195 | 97.4 | 19.5 |
| 0.001 | 7.18 | 48.7 | 24.4 | 4.9 | 48.7 | 24.3 | 4.87 |
| 0.0025 | 280 | 7.81 | 3.90 | 0.78 | 7.79 | 3.90 | 0.779 |
| 0.005 | 4485 | 1.96 | 0.98 | 0.20 | 1.95 | 0.974 | 0.195 |
| 0.01 | 71763 | 0.495 | 0.247 | 0.049 | 0.487 | 0.243 | 4.87E-02 |
| 0.025 | 2.80E+06 | 8.29E-02 | 4.14E-02 | 8.29E-03 | 7.79E-02 | 3.90E-02 | 7.79E-03 |
| 0.05 | 4.49E+07 | 2.30E-02 | 1.15E-02 | 2.30E-03 | 1.95E-02 | 9.74E-03 | 1.95E-03 |
| 0.075 | 2.27E+08 | 1.15E-02 | 5.76E-03 | 1.15E-03 | 8.66E-03 | 4.33E-03 | 8.66E-04 |
| 0.1 | 7.18E+08 | 7.36E-03 | 3.68E-03 | 7.36E-04 | 4.87E-03 | 2.43E-03 | 4.87E-04 |
| 0.2 | 1.15E+10 | 2.98E-03 | 1.49E-03 | 2.98E-04 | 1.22E-03 | 6.09E-04 | 1.22E-04 |
| 0.5 | 4.49E+11 | 1.31E-03 | 6.53E-04 | 1.31E-04 | 1.95E-04 | 9.74E-05 | 1.95E-05 |
| 0.7 | 1.72E+12 | 1.04E-03 | 5.20E-04 | 1.04E-04 | 9.94E-05 | 4.97E-05 | 9.94E-06 |
| 1 | 7.18E+12 | 8.35E-04 | 4.17E-04 | 8.35E-05 | 4.87E-05 | 2.43E-05 | 4.87E-06 |
| 2 | 1.15E+14 | 5.68E-04 | 2.84E-04 | 5.68E-05 | 1.22E-05 | 6.09E-06 | 1.22E-06 |
| 10 | 7.18E+16 | 2.49E-04 | 1.25E-04 | 2.49E-05 | 4.87E-07 | 2.43E-07 | 4.87E-08 |

*- particle diameter below Pe = 1; actual settling rates delayed by the effects of Brownian motion.

A “worst-case” scenario in terms of $PuO_2$ particle size distribution is presented by oxide generated when Pu metal is permitted to passively oxidize in air at ambient conditions (Figure 11). Note that this type of oxide is not produced during ARIES operations. The PSD of two different samples of this type of oxide has an almost normal distribution, with mean and median values between 1 and 2 microns. On average, only 4.2 volume % of this material is smaller than 0.62 $\mu$m, and the majority of this material would settle out of a standing water column in less than 48 hours.

## Conclusions

We applied two well-characterized and simple methods, Stokes’ Law and Equation 4 from Ferguson and Church (2004), to estimate settling velocities of $PuO_2$ particles in a static water column. The estimations derived from each model yield finite time spans for the suspension of a wide size range of $PuO_2$ particles in a flooded PF-4 glovebox. Figure 5 shows the results of these calculations for the gravitational settling velocities of solid, impermeable $PuO_2$ particles of varying diameters in a static water column at 25°C. Figure 6 and Table 5 show the calculated settling times for $PuO_2$ particles having nominal diameters from 0.1 $\mu$m (=0.0001 mm) to 10 mm in static water columns 1 m, 0.5 m, and 0.1 m in depth. As in all model calculations, some caveats apply. The possibilities for particle flocculation, hindered settling at high particle concentrations, and the contribution of Brownian forces of water molecules acting on tiny $PuO_2$ particles were considered here. Particle flocculation is unlikely, and hindered settling is likely not significant at the water-to-solids ratio that would likely occur in a flooded PF-4 glovebox, even at loadings >20 kg. Complications to settling rates caused by particles having internal porosity, permeable particles, the proximity of the particles to the glovebox walls, and the occurrence of turbulent flows were not considered here.

The enhanced suspension of particles due to the action of Brownian forces upon very fine $PuO_2$ particles was modeled using well-established physical criteria for water at 298K and $PuO_2$. An estimate of the Péclet number, *Pe*, equal to 1.0 for $PuO_2$ yielded a radius of 0.31 $\mu$m, which gives the particle diameter below which Brownian forces dominate particle motion in a fluid. Thus, $PuO_2$ fines below ~0.62 $\mu$m in diameter will remain in suspension longer than predicted by any gravitational settling model (e.g., Stokes’ Law). The behavior of particles larger than ~0.62 $\mu$m in diameter will be dominated by gravitational settling.

The behavior of real-world $PuO_2$ particulates in a flooded glovebox can be inferred from very extensive particle size distribution (PSD) data on literally hundreds of $PuO_2$ samples. The PSD of different classes of $PuO_2$ powders produced by the ARIES Program indicate that, during a glovebox flooding incident, approximately 1.0 % by volume will remain in suspension longer than ~48 hours, assuming a water column 1 m deep. The majority of particles in $PuO_2$ powders handled within PF-4 gloveboxes (5 to 50 $\mu$m in diameter) will settle out of a 1 m deep water column in less than 2 hours.

These data show that criticality models which assume the indefinite suspension of plutonium oxide particles of all diameters across a given particle size distribution may grossly overestimate the likelihood of a criticality event during glovebox flooding. The data presented here suggest that the $PuO_2$-to-$H_2O$ ratios of the suspended oxide will be outside of the areas of concern for spontaneous criticality events.

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